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Abbreviations:

BMDL₁₀: Benchmark dose (lower confidence limit) 10%

MOE: Margin of Exposure

PAHs: Polycyclic aromatic hydrocarbons

TDI: Tolerable Daily Intake

Abstract

Background: Coal tar and bitumen have been historically used to coat the insides of cast iron drinking water mains. Polycyclic aromatic hydrocarbons (PAHs) may leach from these coatings into the drinking water and form a potential health risk for humans.

Objectives: In this study we estimated the potential human cancer risk from PAHs in coated cast iron mains.

Methods: In a Dutch nation wide study we took drinking water samples at 120 locations over a period of 17 days under various operational conditions, such as undisturbed operation, during flushing and after a mains repair and analysed these samples for PAHs. We estimated the health risk associated with an exposure scenario over a lifetime period.

Results: During flushing PAH levels frequently exceeded drinking water quality standards. After flushing these levels dropped rapidly. After the cut and repair of cast iron mains, PAH levels exceeded the drinking water standards for up to 40 days after the repair in some locations.

Conclusions: The estimated Margin of Exposure for exposure to PAHs through drinking water was above 10,000 for all 120 measurement locations, which suggests that PAH exposure through drinking water is of only low concern for consumer health. However, factors that differ among water systems, such as the use of chlorination for disinfection, may influence PAH levels in other locations.

Introduction

Several polycyclic aromatic hydrocarbons (PAHs) have been associated with the development of a wide range of cancers (Boffetta et al. 1997; WHO 2003). For this reason, many countries have limits in their drinking water standards for a number of PAHs (Drinking Water Decree 2011; The Drinking Water Directive 1998; United States Environmental Protection Agency 2009). In the Netherlands, only 3 out of more than 1,000 routine samples taken between 2006 and 2009 exceeded PAH drinking water quality standards (Versteegh and Dik 2007, 2008, 2009, 2010). In spring 2009, a Dutch water company received odour and taste complaints after flushing cast iron drinking water mains. After analysing water samples, the water company concluded that the complaints may have resulted from the presence of PAHs in the drinking water, which was assumed to have originated from the bitumen coating in the cast iron mains. As PAHs are seldom found in drinking water, this raised questions about the frequency of increased PAH levels, the circumstances in which this can occur, and the potential implications for human health.

Cast iron drinking water mains are susceptible to corrosion (van den Hoven and van Eekeren 1988) and the resulting corrosion products can lead to a reduction in drinking water quality and a potential increase in customer complaints (Slaats and Rosenthal 2001). Historically, substances such as coal tar or bitumen were used to coat the insides of iron water pipes, which reduces corrosion by forming a water resistant barrier between the pipe wall and the flow of water (Maier 1998; Miller et al. 1982; van den Hoven and van Eekeren 1988). This practice was used from around 1900 until it was phased out in the 1970s. Since then, metal pipes are coated with cement linings.

Both coal tar and bitumen are highly viscose liquids that include a complex mixture of polycyclic aromatic compounds (Zander 1995). The exact composition of coal tar and bitumen can vary depending on the source of coal or petroleum and the processing techniques used to produce them (Read and Whiteoak 2003). The relatively hydrophobic PAHs have a high affinity for sorption onto the surface of particulate matter (De Maagd et al. 1998; Jonker and Koelmans 2002) and thus are usually found in the natural environment in soils or sediment samples, rather than in water or air (Karlsson and Viklander 2008; WHO 2003). Biofilms can form in water distribution networks under favourable conditions such as in areas with an available nutrient source, stable flow conditions, and a suitable surface or substrate on which to grow (O'Toole et al. 2000; van der Kooij 2002). Maier et al. (2000b) demonstrated that coal tar coatings in drinking water mains support and promote the growth of microbiological colonies to a greater degree than uncoated stainless steel because the coal tar linings act as a substrate and nutrient source for the development and support of biofilm.

Although the purpose of coating cast iron drinking water mains is to protect the cast iron from corrosion, the linings themselves can deteriorate over time. As a result, PAHs can leach into water during low-flow conditions when the contact time between the water and the mains is long (Alben 1980; Bowen et al. 2000; Brandt and de Groot 2001; Maier 1998). In addition, soft, corrosive water can degrade coal tar and bitumen linings, which may lead to the release of PAH containing particulate matter into the water (WHO 2003). Maier (1998) concluded that it is unlikely that diffusion from the linings alone accounts for PAH levels in drinking water, and suggested that leaching could increase when fresh fracture surfaces are created as a result of a water main breakage or invasive maintenance. Damage to coal tar and bitumen linings during invasive maintenance could also cause the release of particulate matter containing PAH (Maier

1998; WHO 2003). Oxidation, due to the presence of dissolved oxygen or chlorine or exposure to UV radiation, is thought to play a part in the hardening of bitumen, which makes the coating more susceptible to damage (Read and Whiteoak 2003), especially during invasive interventions in the network. Hydraulic disturbances such as flow changes or pressure fluctuations could lead to the resuspension of accumulated sediment (Vreeburg 2010), which may result in elevated PAH levels in drinking water (Maier 1998; Maier et al. 2000b). On the other hand, biofilm may prevent leaching of PAHs from the coatings into the water supply (Maier 1998; Maier et al. 2000a, b), but increased levels of PAHs in water supplied through coal tar lined water mains were observed following the use of chlorine as a disinfectant (Maier et al. 1997; Maier et al. 2000b), presumably because of the breakdown of protective biofilm. Chlorination, deoxygenation, removal of nutrient source, hydraulic disturbances and low temperatures can all result in biofilm disintegration.

To summarize, PAHs may leach from bitumen coatings into drinking water, and erosion of bitumen coatings may also cause the release of coating particles that contain PAHs. In addition, PAHs may be increased when PAH-contaminated sediment particles are resuspended. Thus, PAHs might be expected to increase during periods of stagnant water (resulting in increased leaching due to long exposure times), during flushing (because of sediment resuspension and biofilm destruction) and during maintenance (invasive work on pipes).

The literature on PAHs in drinking water distribution systems is limited, and there is little quantitative information on the presence of PAHs in drinking water. Therefore, in 2010 we carried out an extensive research project in collaboration with all Dutch water companies, which serve a total population of almost 17 million people. PAHs were measured in samples collected at 120 locations under different operational conditions to identify the conditions associated with

increased PAHs in drinking water, and estimate the potential human health impact of the resulting PAH exposures.

Material and Methods

Measurement Locations

The Dutch drinking water distribution system has a total length of 112,000 km, of which approximately 10% is cast iron mains installed before 1980. We selected 120 measurement locations throughout the Netherlands with cast iron and steel mains of 80 to 120 mm in diameter that were likely to be coated with coal tar or bitumen because they were constructed before 1980. This resulted in approximately 1 sampling location per 100 km of mains. All ten Dutch water companies participated in the study. The number of measurement locations for each water company was determined based on the length of suspect mains in their networks. No further distinctions were made according to water company.

Measurement protocol

Drinking water samples in cast iron mains were collected by the drinking water companies in their own districts under well defined conditions, such as undisturbed operations, during and directly after flushing, and after the removal of a piece and repair of the water main (Table 1). Two of three samples collected during undisturbed operations were taken with some valves closed to ensure a single feed and a well-defined flow direction. Samples obtained during flushing were taken directly from water hydrants. All other samples were obtained from taps located on the distribution mains that were installed by the water companies prior to the

measurement period. For practical reasons, we did not collect residential tap water samples because access could not be guaranteed at all sampling times.

The water companies sampled each measurement location eight times over a two and a half week period, including six times at a tap, and twice at a hydrant (Table 1). For low- and high-flush samples collected from hydrants, flushing flows corresponding to estimated velocities of 0.35 m/s and 1.0 m/s, respectively, were maintained using a specially constructed calibre plate or by measuring the flushing flow. The flush samples were not taken as part of a cleaning programme designed to remove all particles in a systematic way (Vreeburg 2010). During the flushing actions (day 8), the flow (in m³/h) and pressures were recorded. We calculated the actual flushing flow velocities from the measured flow, pressure and the effective diameter and wall roughness as determined from the removed pieces of the mains. On day 15 the water companies removed a 10–50 cm segment of the cast iron main upstream of each sampling location, and subsequently repaired the main by replacing the removed segment with PVC pipe. The drinking water companies took repeat samples at the locations where the drinking water standards were exceeded (sum of 10 PAHs, see Table 2) until no exceedances were found. This was done according to the Drinking Water Decree and was outside of our measurement protocol. This means that the time between the day 17 sample and the repeat sample varied between water companies. We did use the information to determine the duration of PAH exceedances.

Each water company sent their samples to one of the five accredited laboratories in the Netherlands. These laboratories analysed each sample for 16 EPA priority PAHs (ATSDR 1995), pH, total organic carbon, electric conductivity, temperature, and turbidity. Four laboratories determined the PAHs with high performance liquid chromatography with fluorescence detection after solid phase extraction based on NEN-ISO 7981-2:2005 standard (NEN-ISO 2005); the fifth

laboratory used GC-MS after liquid-liquid extraction using on their own internal methodology for the analysis of semi-volatile compounds. The detection limits varied from 0.005 µg/L to 0.05 µg/L between laboratories and between PAHs. Round robin tests were conducted in 2010 to determine the interlaboratory variability in PAH measurements; the average standard deviation was equal to 0.017 µg/l (Asgadaouan 2010).

We measured the inside diameter, wall thickness, wall roughness (in mm), and effective remaining diameter with a caliper, and visually assessed the coating of each of the 120 cast iron segments that were removed from the sampling locations. When the visual assessment was inconclusive (in approximately 35% of samples), we used a burner to heat the main and did a scent-based assessment to determine the presence and type of coating. We categorized the type of coating as “no coating”, “bitumen coating”, “coal tar coating” or “unidentified” (i.e. there is a coating, but there was no certainty on it being coal tar or bitumen); the thickness of the coating as thin (< 1 mm) or thick; and the coverage of the main as none, less than half, more than half and full (indicated in short as “0%”, “>0-50%”, “>50-<100%” or “100%”).

Health risk assessment

The main outcome of concern for exposure to PAHs is cancer. Risk assessment followed the method used by EFSA to evaluate the risk of PAH contamination in food (EFSA 2008), which is based on the Margin of Exposure (MOE)-approach as recommended for genotoxic carcinogens in food (EFSA 2005). MOE is the ratio between a predefined dose and the estimated human intake. Usually the reference dose is the BMDL₁₀ (BenchMark Dose, Lower confidence limit), that is, the lower 95% confidence limit estimated for the dose that causes no more than a 10% increase in cancer incidence in rodents. A genotoxic carcinogen is considered to be of “low

concern for public health and a low priority for risk management actions” if the MOE for the $BMDL_{10}$ is $\geq 10,000$ (EFSA 2005). EFSA concluded that the sum of 8 PAHs (benz[*a*]pyrene (BaP), chrysene, benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*ghi*]perylene, dibenz[*ah*]anthracene and indeno[1,2,3-*cd*]pyrene) can be used as indicators of the carcinogenic potency of PAH mixtures. The sum of 4 PAHs (PAH4: BaP, chrysene, benz[*a*]anthracene, benzo[*b*]fluoranthene), the sum of 2 PAHs (PAH2: BaP and chrysene), and the concentration of BaP alone (BaP), are also used to assess carcinogenicity by EFSA (EFSA 2008).

For each water sample, we calculated the concentration of each of the 16 individual PAHs, and the summed PAHs (PAH2, PAH4 and PAH8). For each study location, we multiplied the PAH levels in each sample by a worst-case estimate of the duration of exposure to that sample (Table 1) and summed the results to derive an estimate of the average exposure per year in each location, which was assumed to be constant over an individual's lifetime. We assumed that flushing and mains repair would occur once per year, with the duration of exposure to levels measured during flushing or repair determined by the maximum duration of PAH exceedances in this study. The number of mains repairs in cast iron mains is reported by the Dutch water companies as 0.05 to 0.42 per km per year. The highest reported values include mains repairs under pressure, i.e. without taking out a part of the main. With 112,000 km of cast iron mains and 7.3 million household connections the average exposure to cast iron mains repairs in the Netherlands is well below once per year. The duration after repair was determined by the fact that we found a PAH exceedance at day 57 (Table 2) which is 40 days after the sample of repair 2. Although consumers generally do not drink water obtained from hydrants during flushing, we used measured values in these samples to estimate exposures during flushing as a worst-case

scenario. For oral intake, we assumed an average body weight of 60 kg and water intake of 2 litre per day (WHO 2011).

We calculated the MOEs for each of the marker PAH combinations by dividing the specific BMDL₁₀ values derived by EFSA (2008) by the estimated average annual exposure across all 120 locations. The BMDL₁₀ values for BaP, PAH2, PAH4, and PAH8 were 0.07, 0.17, 0.34 and 0.49 mg/kg bw/d, respectively (EFSA 2008). To evaluate non-carcinogenic effects of PAHs, we compared the estimated daily intake of the sum of all PAHs (PAH16) with the Tolerable Daily Intake (TDI) of 30 µg/kg bw/d derived by Baars et al. (2001) for PAHs containing less than 17 hydrogen atoms per molecule (benzo(g,h,i)perylene), which is more stringent than the TDI of 40 µg/kg bw/day for non-carcinogenic PAHs with 17 to 35 hydrogen atoms (anthracene, fluorine, naphthalene and phenanthrene). The TDI represents an estimate of the daily intake level to which humans can be exposed during their entire lifetime without causing adverse health effects.

In addition to oral intake, we used the software model ConsExpo (RIVM 2010) to estimate inhalation exposure during showering, taking into consideration both aerosol formation and evaporation of PAHs. We did not estimate dermal exposure during bathing or showering, as data on dermal absorption of PAH from aqueous matrices are scarce, absorption of PAH is highly dependent on the vehicle used in the tests, and available information suggests that exposure from dermal absorption would be negligible compared with oral ingestion from drinking water. Specifically, Moody and Chu (1995) determined dermal absorption for ¹⁴C-labeled phenanthrene *in vitro* in human skin. Absorption in 24 hours from a 0.1 ml of 1 µg/ml phenanthrene solution on 0.64 cm² skin was 24%. From this, an equilibrium constant in terms of partial pressure (Kp) of 1.6x10⁻³ cm/h was estimated. Based on a maximal phenanthrene

concentration of 6 µg/l in drinking water (Table 2), and a surface body area of 18,000 cm², we calculated that half an hour daily bathing or showering would result in a dermally absorbed dose of only 86 ng/person/day, compared with our estimate of 12,000 ng phenanthrene/person/day from daily oral ingestion of 2 L of drinking water. Data from other studies indicate that dermal absorption of other PAHs is not significantly higher than absorption of phenanthrene (Moody and Chu 1995; Sartorelli et al. 1999; Van Rooij et al. 1995).

Results and discussion

PAH levels and relation with different measurement circumstances

We observed each of the 16 PAHs in at least one of the 960 samples. Of the PAHs measured, naphthalene, phenanthrene, pyrene and fluoranthene were the most prevalent. Fluoranthene was detected more often than any other PAHs in the low- and high-flush samples, and phenanthrene was detected more often in samples collected after main repair (repair 1 and repair 2) (Blokkeer et al. 2010).

During normal operation (undisturbed samples 1, 2, and 3) PAHs were undetectable or were present at very low levels only (Figure 1), consistent with expectations given that PAH drinking water standards are rarely exceeded in the Netherlands. This suggests that leaching of PAHs from bitumen coatings is not likely to lead to elevated PAH levels in drinking water during normal operation.

Flushing samples taken at the hydrants contained the highest PAH levels (Figure 1), which suggests that disturbance of sediment in cast iron mains may lead to high PAH levels, and that the PAHs measured in these samples are associated with particles in the distribution network. PAH levels also were elevated in samples collected after main repair, which supports

the hypothesis that erosion of coating particles, in this case due to mains breakage, leads to increased PAH levels. The duration of raised PAH levels after repair varied: of the 48 repair 1 samples with PAH levels that exceeded the drinking water quality standard of $0.1\mu\text{g/L}$ for the sum of 10 PAHs (listed below Table 2), PAHs were undetectable in 13 samples 2 days later (repair 2), in 28 samples 30-40 days after repair, and in 3 samples taken 42, 44, and 57 days after repair respectively. Five of the 48 locations with elevated PAHs after repair were not resampled after the repair 2 sample was taken.

As PAHs are hydrophobic we expected adsorption to sediment particles, and high PAH levels in the flushing samples were consistent with this expectation. However, PAH levels were not correlated with turbidity (an indication of the amount of suspended particles in the sample) or with estimated flushing velocities (which are associated with sediment resuspension) (data not shown). In addition, PAH levels were not correlated with total organic carbon, pH, electric conductivity, or temperature ($R^2 < 0.2$; data not shown).

Relation between PAH and coating

Of the 120 inspected main parts, 104 had a coating (Table 3). For the remaining 16 parts we could not determine if the main was coated at the time of installation. The scent-based assessment could reveal that even if the current coverage was none, the main originally did have a coating. Of the 104 mains parts with an established coating, we assessed the coating type and coverage (Table 3). We found bitumen, coal tar, and some unidentified coating types of varying thicknesses and coverage percentages (Figure 2). Even in several of the 16 locations with undetermined presence of a coating, PAHs were detectable during flushing (data not shown) and after mains repair (Figure 3). In these cases it is possible that although the segment that was

removed had no coating, the rest of the cast iron main may have been coated. Alternatively, the PAHs that were found may have originated from another part of the network.

This study showed differences in observed PAH levels between the different types of coating (Figure 3). Coal tar coatings were associated with higher mean PAH levels than bitumen coatings (0.63 µg/L; 95% CI: 0.11, 1.16 versus 0.13 µg/L; 95% CI 0.05, 0.21, respectively). Since water companies do not know which of their cast iron mains have bitumen coatings and which have coal tar coatings, the observation of the effect of coating type is of no practical value.

Main segments that were coated with >0 to 50% coverage had the highest mean PAH levels in water samples taken immediately after the segment was removed (Figure 3) though a one-way analysis of variance showed no differences among mean PAH concentrations according to the amount of coverage. Mains with >0–50% coverage also had higher mean PAH levels in the flush samples (data not shown). These findings support the hypothesis that coating erosion may contribute to increased PAH levels. The year the main was installed was not a significant predictor of the level of coating coverage (data not shown).

Human health risk

We compared the PAH levels in the water samples to the drinking water quality standards of 0.1 µg/l for the sum of 10 PAHs and 0.01 µg/L for benzo[*a*]pyrene (Drinking Water Decree 2011). Under normal operation (undisturbed 1 and 2 samples) no exceedances of PAH standards were observed (Table 2). However, PAH levels in some samples collected shortly after flushing and repair exceeded the total PAH (29% and 40% of samples, respectively) and BaP standards (24% and 6%), and 31% of samples collected two days after repair exceeded the total PAH standard.

An exposure with an MOE of 10,000 or higher is considered to be of low concern for public health and a low priority for risk management actions (EFSA 2005). MOEs calculated for oral exposure in a 60 kg adult consuming 2 litres of water per day based on the maximum PAH8 concentration measured in any sample of a given type were <10,000 for low- and high-flush samples, samples taken 15 minutes after flushing, and samples taken two to four hours after repair (Table 2). However, these MOEs were based on the highest measured level, and the test conditions with MOEs <10,000 are likely to be uncommon within a distribution network. Therefore, we calculated MOEs for carcinogenic effects at each individual location after estimating annual oral intakes based on the BaP, PAH2, PAH4, and PAH8 concentrations in the 8 routinely collected samples from each location after accounting for estimated exposure durations for each sample type (specifically, 1 day of exposure to the PAH concentration measured in the low-flush sample, 1 day for the high-flush concentration, 2 days for the after flush, 2 days for repair 1, 40 days for repair 2, and 319 days of exposure to the maximum concentration measured in the 3 samples collected during undisturbed operation, Table 1). Because we included exposures based on concentrations in flush samples collected from hydrants, which are not normally consumed, we considered these intake estimates to be conservative (i.e., higher than would be expected for most consumers.) All MOEs estimated for the 120 locations using this method were above 10,000, with the lowest estimated MOE = 104,000 (Figure 4).

Estimates of inhalational exposure derived using ConsExpo (RIVM 2010) and assuming the maximum concentration measured in any sample for each individual PAH indicated that inhalation exposure by aerosol formation during showering was negligible compared with estimated oral intakes (<1% of oral exposure for all 16 PAHs combined; data not shown),

whereas inhalation exposure due to evaporation of volatile PAHs was comparable to oral exposure for the sum of the maximum concentration measured for each of the 16 PAHs (2.0 and 2.1 $\mu\text{g/kg bw/day}$, respectively). However, only 0.2, 10, 11 and 6.9% of the total exposure to the carcinogenic PAHs (BaP, PAH2, PAH4, and PAH8 respectively) was attributed to inhalation during showering, which suggests that inhalatory exposure primarily involves PAHs classified as non-carcinogens, or as low potent carcinogens. Therefore, we did not include inhalation exposure during showering when doing risk assessment for carcinogenic effects of PAHs in drinking water.

Toxic effects other than carcinogenic effects are not likely to occur, as exposure to the sum of the maximum concentrations of each of the 16 PAHs measured, considering both oral and inhalatory routes of exposure, was determined at 4.1 $\mu\text{g/kg bw/day}$ (Table 4), whereas the lowest derived TDI for non-carcinogenic effects of exposure during a single day is 30 $\mu\text{g/kg bw/d}$ (Baars et al. 2001).

General discussion

Exceedance of PAH standards can occur after invasive repairs in cast iron mains, but our estimates suggest that human health risks related to coated cast iron water mains are low. To prevent increased exposures during flushing operations, drinking water networks should be flushed in a systematic way to ensure that all particles with PAH are removed (Vreeburg 2010).

Water quality aspects such as hardness (WHO 2003), chlorination (Maier et al. 1997; Maier et al. 2000b), and oxidation (Read and Whiteoak 2003) may affect PAH levels in drinking water. Chlorination is not used to disinfect drinking water in the Netherlands, and we did not evaluate hardness and oxidation, which have varied over time. We also did not assess the influence of biofilm on PAH levels as suggested by Maier et al. (1997, 2000b).

PAH levels also are likely to be increased in cast iron mains as a consequence of invasive work and raised flow velocities in water distribution networks outside of the Netherlands. However, the specific circumstances associated with elevated PAH levels may differ, for example, in countries where chlorine is used as a disinfectant, and additional field measurements would be needed to determine whether our findings apply elsewhere.

Conclusions

PAHs from coal tar and bitumen coatings in cast iron water mains are a potential health risk for humans. Over a period of 17 days at 120 measurement locations in cast iron distribution networks in the Netherlands, we took drinking water samples under various operational conditions, including undisturbed operation, during flushing, and after repair of water mains. We analysed these samples for PAHs and estimated the health risk by the MOE-approach. During flushing high PAH levels may occur, but after flushing PAH concentrations dropped rapidly. After coated cast iron mains were cut and repaired we measured PAH levels that exceeded the water quality standard up to 40 days after the repair. However, even after accounting for such exceedances, estimated MOEs were above the 10,000 benchmark for all 120 measurement locations. Therefore, we conclude that PAH exposure through Dutch drinking water is of only low concern for consumer health.

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Table 1 Sampling times, sample conditions, and estimated exposure durations

Day	Sample	Conditions	Exposure Duration (Days)
1	undisturbed 1	sample taken at tap during undisturbed normal operation	
3	undisturbed 2	sample taken at tap during undisturbed operation on single feed	
8	low-flush	sample taken at hydrant during low velocity flush (0.35 m/s), 15 s after opening hydrant	1
8	high-flush	sample taken at hydrant during high velocity flush (1.00 m/s), 15 s after opening hydrant	1
8	after-flush	sample taken at tap after 15 minutes after hydrant was closed during undisturbed operation (on single feed)	2
10	undisturbed 3	sample taken at tap during undisturbed operation (on single feed)	319*
15	repair 1	sample taken at tap during undisturbed operation (on single feed) after two to four hours after pipe was closed after the removal and repair of a piece of cast iron main	2
17	repair 2	sample taken at tap after 2 days after pipe was closed during undisturbed operation (on single feed)	40

* The maximum value of the three samples during undisturbed operation was assumed to occur the rest of the year, i.e. 319 days.

Table 2 PAH exceedances in samples according to Dutch Drinking Water Decree (Drinking Water Decree 2011) assuming the maximum concentrations measured in any sample of a given type (120 samples per type).

sample	exceedance of PAH drinking water quality standard (0.1 µg/l for the sum of 10 PAH) ^a	exceedance of benzo[<i>a</i>]pyrene drinking water quality standard (0.01 µg/l)	Maximal sum PAH8 (in µg/l)	Maximal oral exposure PAH8 (µg/kg bw/day)	Margin of Exposure PAH8
Undisturbed 1	no	no	0.003	0.0001	4,900,000
Undisturbed 2	no	no	0.001	0.00003	14,700,000
Low-flush ^b	60% of samples	49% of samples	19	0.62	794
High-flush ^b	60% of samples	61% of samples	22	0.75	656
After-flush	29% of samples	24% of samples	2.1	0.069	7,067
Undisturbed 3	2% of samples	no	0.020	0.0007	735,000
Repair 1	40% of samples	6% of samples	2.0	0.069	7,313
Repair 2	31% of samples	2% of samples	0.23	0.0077	63,913

^a Sum of concentration of anthracene, benz[*a*]anthracene, benzo[*b*]fluoroanthene, benzo[*ghi*]perylene, benzo[*k*]fluoroanthene, chrysene, fluoroanthene, indeno[1,2,3-*cd*]pyrene, phenanthrene, and pyrene.

^b Samples taken at hydrant.

Table 3 Coating characteristics of 120 pipes.

Coating characteristic		Number of pipes	Percentage of pipes^a
Coated	Yes	104	87
	Undetermined	16	13
Coating type	Bitumen	60	58
	Coal tar	21	20
	Unidentified	23	22
Thickness	Thin	70	67
	Thick	34	33
Coverage	0%	9	9
	>0-50%	36	35
	>50-<100%	27	27
	100%	32	31

^aPercentage of all samples for Coated (n = 120), otherwise percentages are for all coated pipes (n = 104)

Table 4. Estimated oral and inhalation exposures to measured and summed PAHs on day of sampling ^a

PAH	max conc. (µg/l)	Exposure (µg/kg bw/d)	
		Inhalation	oral
acenaphtene	0.96	0.082	0.032
acenaphtylene	0.64	0.055	0.021
anthracene	1.7	0.14	0.057
benz[<i>a</i>]anthracene	4.3	0.0093	0.14
benzo[<i>b</i>]fluoranthene	3.8	0.024	0.13
benzo[<i>k</i>]fluoranthene	2.2	0.0000048	0.073
benzo[<i>ghi</i>]perylene	2.6	0.0000057	0.087
benzo[<i>a</i>]pyrene	3.6	0.00027	0.12
chrysene	3.6	0.028	0.12
dibenz[<i>ah</i>]anthracene	1.8	0.0000053	0.060
fluoranthene	11	0.32	0.37
fluorene	1.4	0.12	0.047
indeno[1,2,3- <i>cd</i>]pyrene	2.7	0.0000053	0.090
naphtalene	7.4	0.63	0.25
phenanthrene	6	0.51	0.20
pyrene	6.6	0.16	0.22
PAH2		0.028	0.24
PAH4		0.061	0.51
PAH8		0.061	0.82
PAH16		2.1	2.0

^a Estimates are based on the maximum concentration measured in any sample..Summed PAHs are theoretical values as they are composed of the maximum concentrations measured for the individual PAHs included in different samples.

Figure Legends

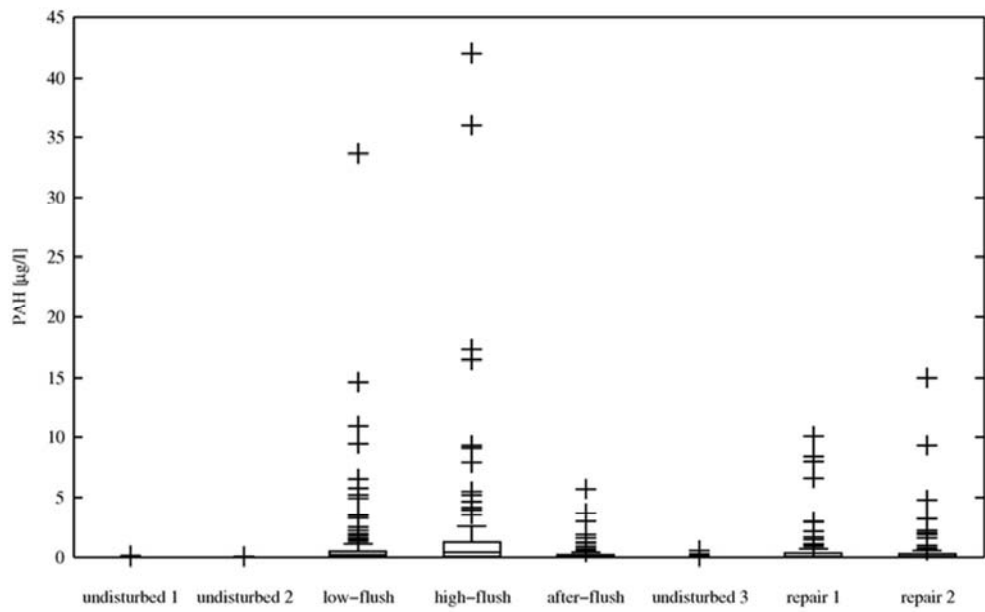
Figure 1. Box and whisker plot of the sum of 16 PAH in each sample type (n = 120 for each).

Boxes extend from the 25th to the 75th percentile, horizontal bars represent the median, whiskers extend 1.5 times the length of the interquartile range (IQR) above and below the 75th and 25th percentiles, respectively, and outliers are represented as points.

Figure 2. Photographs showing coating of (A) location 104: 0% coal tar coating; (B) location 41: >0-50% bitumen coating (thin); (C) location 83: >50-<100% bitumen coating (thick); (D) location 34: 100% bitumen coating (thick).

Figure 3. Box and whisker plot of PAH concentrations in samples taken 2–4 hours after a water main segment was removed upstream of the sampling location according to a) type of coating and b) coating coverage. Numbers of samples are indicated in parentheses. Boxes extend from the 25th to the 75th percentile, horizontal bars represent the median, whiskers extend 1.5 times the length of the interquartile range (IQR) above and below the 75th and 25th percentiles, respectively, and outliers are represented as points.

Figure 4. Margins of Exposure for exposure to carcinogenic PAHs (BaP, PAH2, PAH4, and PAH8) through drinking water at all 120 measurement locations. Annual oral intakes were estimated based on concentrations in the 8 routinely collected samples from each location after accounting for estimated exposure durations for each sample type. MOEs for samples with no measurable PAHs are infinite and therefore are not shown in the graph.



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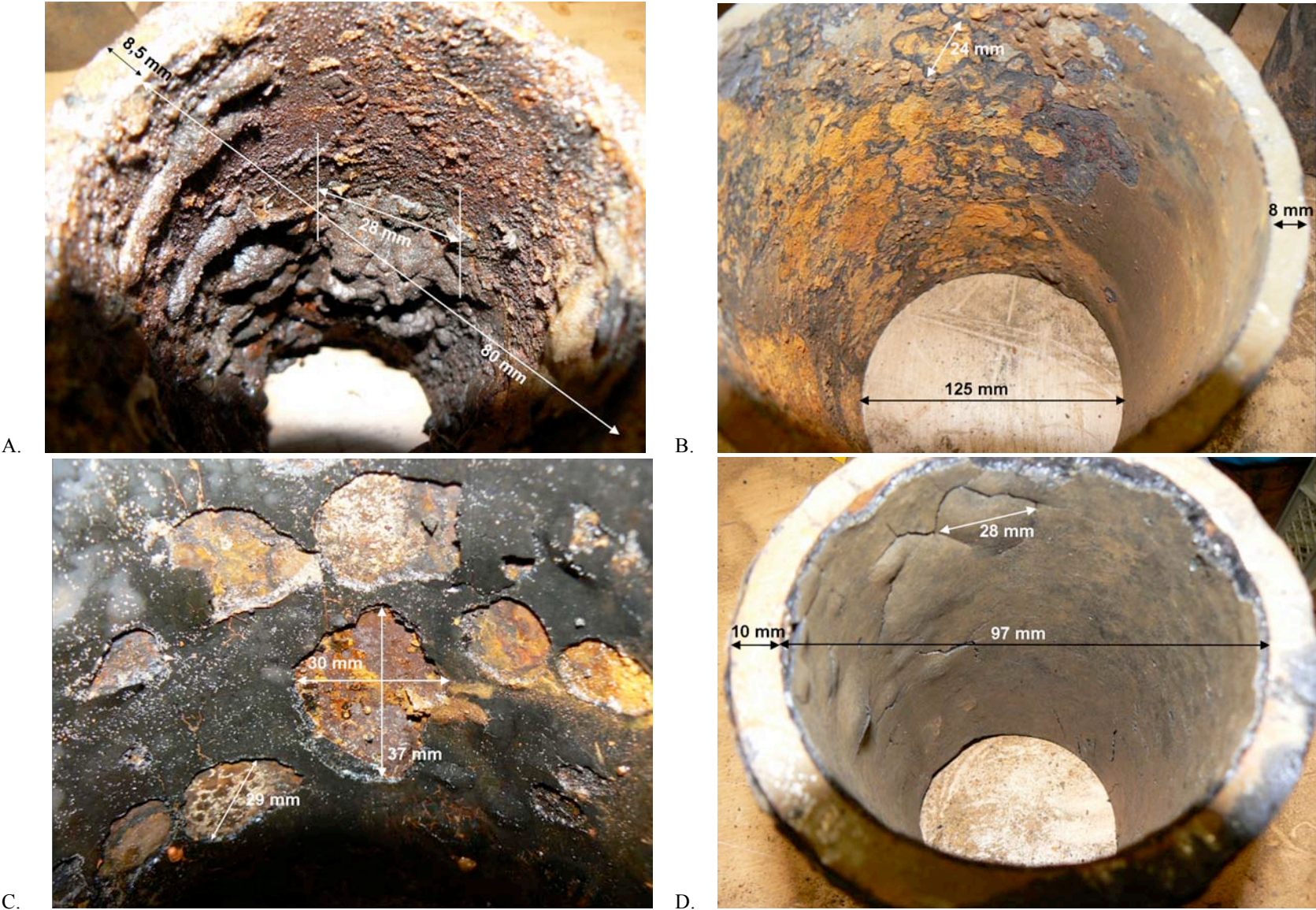
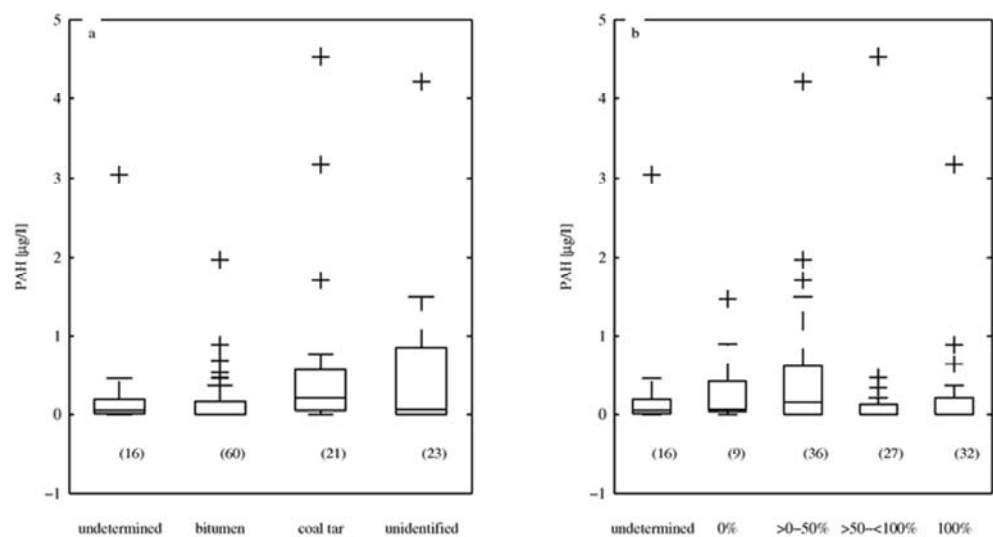
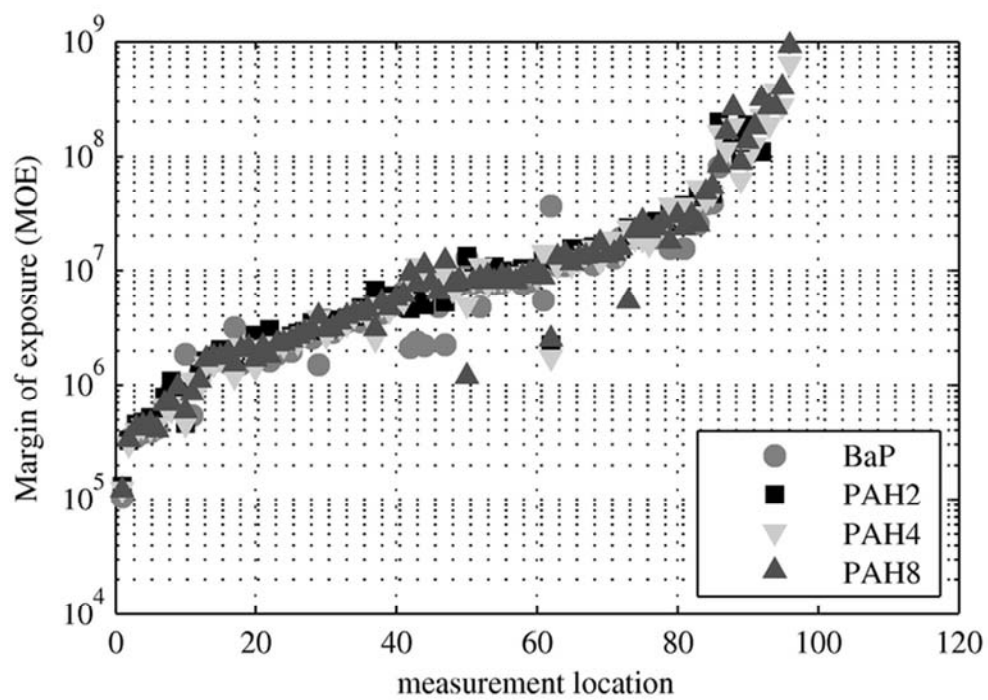


Figure 2



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